Impurity scattering in superconductors[†]

Yong-Jihn Kim and K. J. Chang

Department of Physics, Korea Advanced Institute of Science and Technology,

Taejon 305-701, Korea

Abstract

We present some of the recent theoretical studies on the impurity effects in conventional superconductors, such as magnetic and ordinary impurity effects in dirty and weak localization limits, and describe successfully unexplained experimental results. We find that the critical sheet resistance for the suppression of superconductivity in thin films depends on superconductor, and point out that impurity dopings in high T_c superconductors cause a metal-insulator transition and thereby suppress T_c.

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I. INTRODUCTION

Although there have been much studies on the impurity doping effects in high T_c superconductors,[1,2] it has not been successful to understand the underlying physics. In particular, the decrease of T_c due to impurity scattering is not fast enough to support the anisotropic pairing model, such as d-wave or anisotropic s-wave pairing. In conventional low T_c superconductors, it was shown that the Abrikosov and Gor'kov's (AG) Green function theory is in conflict with the Anderson's theory of dirty superconductors.[3–5] The Anderson's theory predicted no substantial decrease of T_c with ordinary impurity substitution, while the AG theory leads to a large decrease of T_c, which varies linearly with impurity concentration. For magnetic impurity effects, the T_c reduction caused by exchange scattering was found to be suppressed by adding non-magnetic impurities or radiation damage,[6,7] which contradicts to the AG theory. In weak localization limit, ordinary impurities also decrease T_c, which is beyond the Anderson's theory. Previously, this feature was attributed to the enhanced Coulomb repulsion due to impurities,[8,9] however, tunneling experiments did not show any increase of the Coulomb repulsion.[10,11]

In this paper, we present some of our recent theoretical work[3,12–15] on the impurity effects in conventional superconductors, which describe successfully unexplained experimental results. We first discuss the magnetic impurity effect and show why ordinary impurities and radiation damage suppress the T_c reduction caused by magnetic impurities within the framework of the BCS theory. The ordinary impurity effect is considered both in dirty and weak localization limits. In weak localization limit, where the Anderson's theorem is not valid, T_c is suppressed because of the amplitude modulation of the wavefunction caused by impurity scattering. In two-dimensional samples, it is shown that the critical sheet resistance for the suppression of superconductivity depends on superconducting material, in good agreement with experiments.[16,17] We point out that the impurity doping and ion-beam-induced damage in high T_c superconductors give rise to a metal-insulator transition and thereby suppress T_c.

II. IMPURITY SCATTERING IN CONVENTIONAL SUPERCONDUCTORS

A. Magnetic impurity scattering

Magnetic impurities suppress strongly the transition temperatures of singlet-pairing superconductors. The magnetic interaction between a conduction electron at $\mathbf{r}$ and a magnetic solute at $\mathbf{R}_i$ is given by

$$H_{mag} = \sum_i J\vec{s}\cdot\vec{S}_i\delta(\mathbf{r}-\mathbf{R}_i), \tag{1}$$

where $\vec{s} = \frac{1}{2}\vec{\sigma}$ and $\vec{\sigma}$ represents the Pauli spin operators. Employing the degenerate scattered-state pairs with the magnetic scattering effect, Kim and Overhauser[12] showed that the phonon-mediated matrix elements are expressed as

$$V_{nn'} = -V\langle cos\theta_{n'}(\mathbf{r})cos\theta_n(\mathbf{r})\rangle_{av}, \tag{2}$$

where $\langle\ \rangle_{av}$ denotes the average over $\mathbf{r}, \vec{R}_i$ and the spin direction $\vec{S}_i$ of the solutes. Here $cos\theta(\mathbf{r})$ is the relative singlet amplitude of the basis pair when both the electrons are near $\mathbf{r}$. In the case of magnetic scattering, the singlet amplitude of the Cooper pair is reduced, and consequently T_c is decreased.

Notice that only the magnetic impurities within the BCS coherence distance ξ_o (for a pure superconductor) from the Cooper pair's center of mass reduce both the singlet amplitude and the pairing interaction. Then, one finds

$$\langle cos\theta\rangle = 1 - \frac{\pi\xi_o}{2\ell_s}, \tag{3}$$

where

$$\xi_o = 0.18\frac{\hbar v_F}{k_BT_c}, \tag{4}$$

and ℓ_s is the mean free path for the exchange scattering only and v_F is the Fermi velocity. Here, we use an iterative method to calculate T_c from the BCS gap equation. The change

of T_c with respect to T_{co} (for a pure metal) can be easily calculated to the first order in impurity concentration, such as

$$k_B \Delta T_c \cong -\frac{0.18\pi\hbar}{\lambda\tau_s}, \tag{5}$$

where λ is the electron-phonon coupling constant and τ_s is the spin-disorder scattering time. Since ΔT_c is inversely proportional to λ, the initial slope (versus $1/\tau_s$) depends on superconductor, indicating that this slope is not the universal constant, while the universal behavior was proposed by the AG theory. As illustrated in Fig. 1, the critical temperatures of weakly-coupled superconductors are found to decrease rapidly with increasing of magnetic impurities, as compared to strongly-coupled superconductors.

For the conduction electrons with a mean free path ℓ, which is smaller than ξ_o, the effective coherence length is reduced to

$$\xi_{eff} \approx \sqrt{\ell \xi_o}. \tag{6}$$

Then, the reduction of T_c by magnetic impurities is significantly suppressed because the number of magnetic impurities within the distance of ξ_{eff} is very small. This behavior was first observed by co-doping with non-magnetic impurities that result in the decrease of ℓ.[6] A similar compensation effect was also observed by radiation damage; for pure and Mn-implanted In-films, the change of T_c were found to be 2.2 and 0.3 K, respectively, after an exposure to a 275 kev Ar^+-ion fluence of $2.2\times10^{16} cm^{-2}$.[7] This compensation effect contradicts to the traditional belief that ΔT_c in magnetically doped superconductors is unaffected by non-magnetic scatterings.[4]

B. Ordinary impurity scatterings in dirty and weak localization limits

To describe the ordinary impurity effect, Anderson introduced the exact scattered states $\psi_{n\sigma}$ for the conduction electrons in a metal with ordinary impurities, which form time-reversed scattered state pairs. The scattered state $\psi_{n\sigma}$ can be expanded in terms of plane waves $\phi_{\vec{k}\sigma}$, such as

$$\psi_{n\sigma} = \sum_{\vec{k}} \phi_{\vec{k}\sigma} \langle \vec{k} | n \rangle. \tag{7}$$

Then, the phonon-mediated matrix elements between the time-reversed pairs are written as[3]

$$V_{nn'} = -V(1 + \sum_{\vec{k} \neq -\vec{k}', \vec{q}} \langle -\vec{k}' | n \rangle \langle \vec{k} | n \rangle^* \langle \vec{k} - \vec{q} | n' \rangle \langle -\vec{k}' - \vec{q} | n' \rangle^*). \tag{8}$$

Although the correction term in the right hand side of Eq. (8) is negligibly small in dirty limit, it is important in weak localization limit. Consequently, the Anderson's theorem is valid only in dirty limit, to the first order in impurity concentration.[3]

It was shown[13] that the Tsuneto's strong coupling theory[18] fails to explain the existence of the localization correction in the phonon-mediated interaction. Alternatively, from the real space formalism of the strong coupling theory with the time-reversed pairs, Kim[13] obtained a strong coupling gap equation

$$\Delta(n, \omega) = \sum_{\omega'} \lambda(\omega - \omega') \sum_{n'} V_{nn'} \frac{\Delta(n', \omega')}{\omega'^2 + \epsilon_{n'}^2}, \tag{9}$$

where

$$V_{nn'} = -V \int |\psi_n(\mathbf{r})|^2 |\psi_{n'}(\mathbf{r})|^2 d\mathbf{r}, \tag{10}$$

$$\lambda(\omega - \omega') = \frac{\omega_D^2}{\omega_D^2 + (\omega - \omega')^2}, \tag{11}$$

and ω_D is the Debye frequency. Here ω' and $\epsilon_{n'}$ represent the Matsubara frequency and the electron energy, respectively. Using the wavefunction in Eq. (7), one can easily derive the formula of Eq. (8) from Eq. (10). We point out that $V_{nn'}$ gives the change of the phonon-mediated interaction due to impurities, and it decays exponentially for the localized states. In the Tsuneto's theory, however, it remains unchanged, even if the wavefunctions are localized.

For the strongly localized states, since both the phonon-mediated interaction and the conductivity decay exponentially, they are expected to have the same correction term in weak localization limit. Kaveh and Mott[19] showed that the wavefunction for the weakly localized

states may be written as a mixture of power-law and extended wavefuctions. Employing their wavefunctions and considering only the impurities within the distance of ξ_{eff}, the following relations for the matrix elements were obtained;[14,15]

$$V_{nn'}^{3d} \cong -V[1 - \frac{3}{(k_F\ell)^2}(1 - \frac{\ell}{L})], \tag{12}$$

$$V_{nn'}^{2d} \cong -V[1 - \frac{2}{\pi k_F \ell} ln(L/\ell)], \tag{13}$$

$$V_{nn'}^{1d} \cong -V[1 - \frac{1}{(\pi k_F a)^2}(L/\ell - 1)], \tag{14}$$

where ℓ and L are the elastic and inelastic mean free paths, respectively, and a is the radius of an one-dimensional wire.

Solving the BCS gap equation with the matrix element of Eq. (12), the change of T_c with respect to T_{co} satisfies the relation

$$\frac{T_{co} - T_c}{T_{co}} \propto \frac{1}{(k_F\ell)^2}, \tag{15}$$

for bulk materials and this result is in good agreement with experiments,[20] as shown in Fig. 2. In homogeneous two-dimensional thin films, an empirical formula was obtained,[21]

$$\frac{T_{co} - T_c}{T_{co}} \propto \frac{1}{k_F\ell} \propto R_{sq}, \tag{16}$$

where R_{sq} denotes the sheet resistance, and in fact this formula can be derived by putting Eq. (13) into the BCS gap equation. Previously, the decrease of T_c due to disorder was attributed to the enhanced Coulomb repulsion,[8,9] whereas tunneling measurements do not support this picture.[10,11] The dirty boson theory[22] predicted the universal critical sheet resistance of $R_q = h/4e^2 = 6.4k\Omega$ for the suppression of superconductivity in thin films. However, we find that from Eq. (13) the critical sheet resistance is not universal but sample-dependent, which agrees with experiments.[17,23]

III. IMPURITY SCATTERING IN HIGH T_C SUPERCONDUCTORS

There is currently considerable interest in the symmetry of the superconducting state in high T_c superconductors.[24] Impurity doping studies may give a clue of resolving this

problem. Since the normal-state transport behavior of the cuprates is so anomalous, the impurity doping effects on the superconducting state may not be easy to understand. We note that almost all the experiments with Cu ions substituted by other metal ions show the metal-insulator transition driven by impurity dopings.[25,26] Thus, the decrease of T_c seems to be closely related to the wavefunction localization. If the d-wave pairing and the Fermi liquid theory are assumed in the cuprates, since T_c decreases much faster with increasing of impurities,[12] the superconductivity may disappear before the metal-insulator transition is reached. In ion-beam irradiation and ion implantation experiments[27,28] as well as $Y_{1-x}Pr_xBa_2Cu_3O_7$ samples,[29,30] the metal-insulator transitions were also found in the doping region, where T_c drops to zero. This anomalous impurity doping effect seems to imply that the Landau's quasi-particle picture is not applicable for the cuprates. To understand this anomalous behavior, we may need to understand the impurity effect on the normal state.

Recently, Suryanarayanan et al.[31] found the recovery of superconductivity in $Y_{1-x}Ca_xSrBaCu_{2.6}Al_{0.4}O_{6+z}$ when Y is substituted by Ca; the values of T_c were found to be 0, 29, and 47 K for x = 0, 0.1 and 0.2, respectively. This results may be understood if we consider the importance of the metal-insulator transition caused by impurity doping. When x = 0, the increase of Al impurities changes the system to the insulating state with the mobility edge lying below the Fermi energy. Then, since the conducting electrons are localized, the superconductivity transition does not appear. If holes are added in the Cu-O planes with Ca impurities, since the Fermi energy moves below the mobility edge, assuming the mobility edge unchanged, and the electrons are extended, Ca-doped systems become superconducting. Further experimental studies are needed to understand clearly the impurity-driven metal-insulator transition in the cuprates.

IV. CONCLUSIONS

Within the framework of the BCS theory, we have discussed the magnetic and non-magnetic impurity effects on conventional superconductors. In particular, we find that the critical sheet resistance for the suppression of superconductivity in thin films is not universal but sample-dependent. For high T_c superconductors, it is pointed out that the metal-insulator transition driven by impurity doping is important in understanding both the normal and superconducting states of the cuprates.

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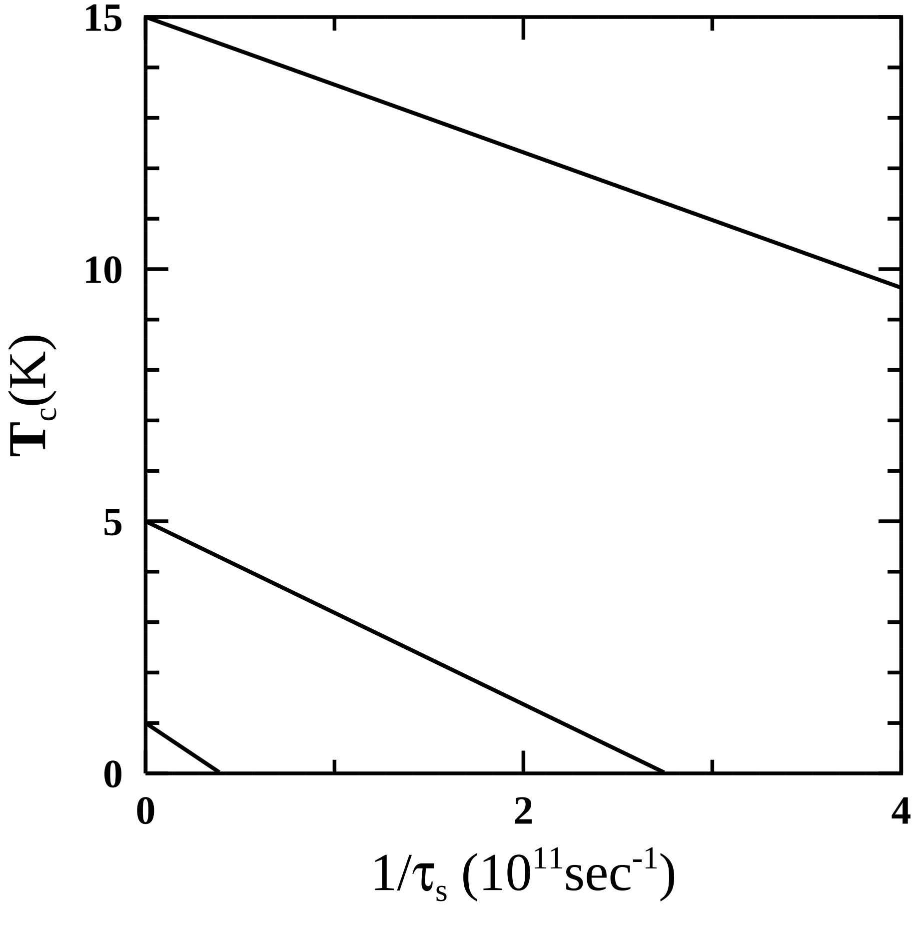
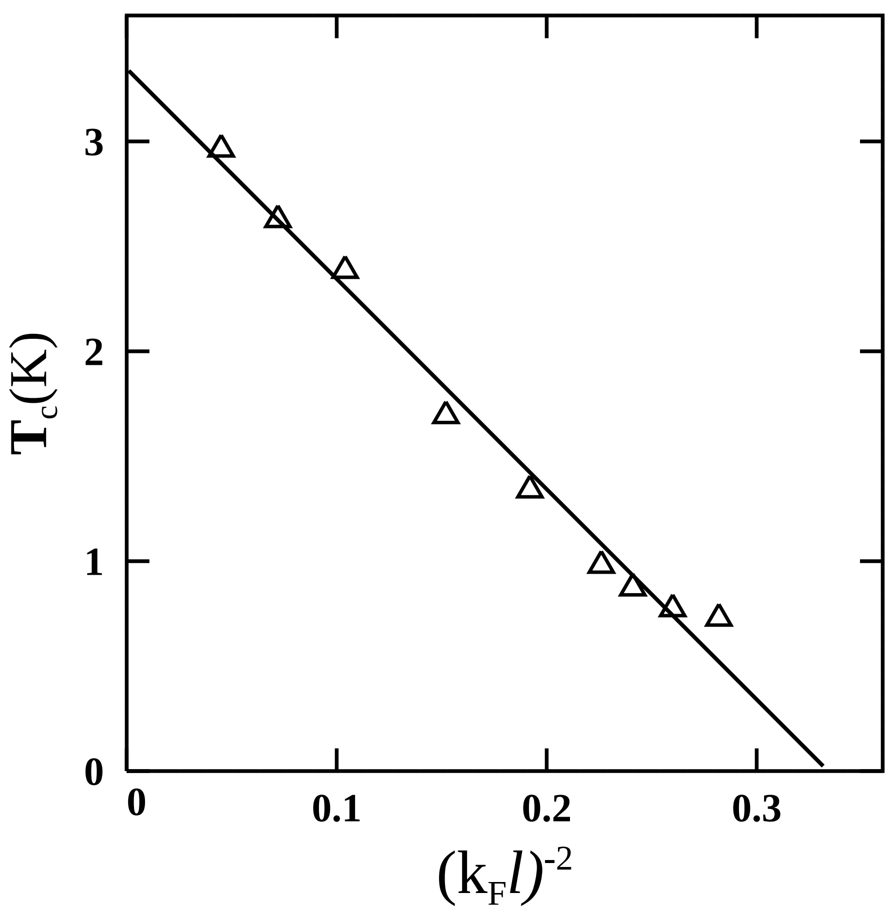
Figure Captions

Fig.1 Initial slopes of T_c with varying the spin-disorder scattering rate $(1/\tau_s)$ for T_c =1, 5, and 15K.

Fig.2 The calculated superconducting temperatures (solid line) of InO_x are plotted as a function of $(k_F\ell)^{-2}$ and compared with experiments (triangles) from Ref. 20.



15
10
5
0
0
2
4
T_c(K)
1/τ_s (10^11 sec^-1)



T_c(K)
3
2
1
0
0
0.1
0.2
0.3
$(k_F l)^{-2}$